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Interrelation between the pseudogap and the incoherent quasi-particle features of high-T_c superconductors

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Using a scenario of a hybridized mixture of localized bipolarons and conduction electrons, we demonstrate for the latter the simultaneous appearance of a pseudogap and of strong incoherent contributions to their quasi-particle spectrum which arise from phonon shake-off effects. This can be traced back to temporarily fluctuating local lattice deformations, giving rise to a double-peak structure in the pair distribution function, which should be a key feature in testing the origin of these incoherent contributions, recently seen in angle resolved photoemission spectroscopy ($ARPES$).

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The appearance of a pseudogap, accompanied by a predominantly incoherent quasi-particle spectrum in certain parts of the Brillouin zone [1], is considered to be amongst the most significant signatures of high-T_c superconductors (HT_cSC) which may contain the key of our understanding of these materials. As suggested earlier, the large incoherent part of the quasi-particle spectrum might come from a coupling of the electrons to collective modes such as spin fluctuations [2]. We shall discuss and defend in this Letter a similar point of view based on a scenario of a mixture of intrinsically localized bipolarons and coexisting itinerant electrons, hybridized with each other via charge exchange, permitting bipolarons to disintegrate into pairs of conduction electrons and vice-versa to reconstitute themselves in an inverse process. The location of the bipolarons in high-T_c materials might be sought in the highly polarizable dielectric layers adjacent to the CuO_2 planes or possibly inside the polaronic stripes [3] in those planes themselves - the remainder of those CuO_2 planes forming the subsystem housing the itinerant electrons. Taking the bipolarons as quasi-particles without any internal structure, such a scenario is described by the so called Boson-Fermion model (BFM) which has led us to the prediction of pseudogap features in the quasi-particle spectrum [4], driven by strong local electron-pair correlations. In the present Letter we extend our previous studies by taking into account the internal polaronic structure of the bipolaronic Bosons, as being composed of charge and lattice vibrational degrees of freedom, locked together in a coherent quantum state. A bipolaronic Boson localized on a site i is represented by

$$b_i^+ \; e^{-\alpha(a_i - a_i^+)}|0\rangle|\Phi(X)) = b_i^+|0\rangle|\Phi(X - X_0)). \qquad (1)$$

where the phonon operators $a_i^{(+)}$ correspond to local lattice deformations. The hard-core Bose operators $b_i^{(+)}$ describe pairs of electrons which are self-trapped inside locally deformed clusters of atoms, characterized by deformed harmonic oscillator states $|\Phi(X - X_0))$ with equilibrium positions shifted by $X_0 = 2\alpha\sqrt{\hbar/2M\omega_0}$ (ω_0 denotes the characteristic frequency and M the mass of the oscillators). The strength of the coupling of the charge carriers to local lattice deformations, ultimately leading to bipolaron formation, is given by $\hbar\omega_0\alpha$. Such physics is described in terms of the following generalization of the original BFM:

$$\begin{aligned} H = &(D-\mu)\sum_{i,\sigma} c_{i\sigma}^+ c_{i\sigma} - t \sum_{\langle i\neq j\rangle,\sigma} c_{i\sigma}^+ c_{j\sigma} \\ &+ (\Delta_B - 2\mu)\sum_i b_i^+ b_i + v\sum_i [b_i^+ c_{i\downarrow} c_{i\uparrow} + c_{i\uparrow}^+ c_{i\downarrow}^+ b_i] \\ &- \hbar\omega_0\alpha \sum_i b_i^+ b_i (a_i + a_i^+) + \hbar\omega_0 \sum_i \left(a_i^+ a_i + \frac{1}{2}\right). \quad (2) \end{aligned}$$

Here $c_{i\sigma}^{(+)}$ are Fermionic operators referring to itinerant electrons with spin σ. The bare hopping integral for the electrons is given by t, the bare Fermionic half band width by D, the Boson energy level by Δ_B and the Boson-Fermion pair-exchange coupling constant by v. The chemical potential μ is common to Fermions and Bosons. The indices i denote effective sites involving molecular units made out of adjacent molecular clusters of the metallic Fermionic and dielectric Bosonic subsystems. Because of the small overlap of the oscillator wave functions at different sites we may, to within a first approximation, consider the Boson and Fermion operators as commuting with each other.

The original BFM, given by the first two lines in Eq.(2), has been investigated in great detail as far as the opening of the pseudogap is concerned and as far as this affects the thermodynamic, transport and magnetic properties [4]. The opening of the pseudogap in the Fermionic density of states was shown to be driven by

the onset of local electron pairing without any superconducting long range order. Even without treating the generalized BFM within the self-consistent conserving approximation, used in those studies of the original BFM, we find that the atomic limit of this generalized BFM already gives us clear indications on the interrelation between the opening of the pseudogap and the appearance of predominantly incoherent quasi-particle features, as seen in ARPES studies.

In order to set the scale of the various parameters in this model we measure them in units of D, which for typical HT_cSC is of the order of 0.5 eV. As in our previous calculations of the original BFM, we choose v such that the pseudogap opens up at temperatures of the order of a hundred degrees K. We take $v = 0.25$ for the present study. We furthermore choose $\alpha = 2.5$ such that together with a typical local phonon frequency of the order of $\omega_0 = 0.1$ we have a reasonable bipolaron binding energy $\varepsilon_{BP} = \alpha^2\hbar\omega_0$ which pins the chemical potential at about half the renormalized Bosonic level $\tilde{\Delta}_B = \Delta_B - \hbar\omega_0\alpha^2$. We choose $\tilde{\Delta}_B$ to lie close to the band center such that the number of electrons is slightly below half-filling (typically around 0.75 per site, which is the physically relevant regime of concentrations). For larger binding energies the bipolaronic level would drop below the band of the electrons leading to a situation of *bipolaronic superconductivity*, which is clearly not realized in HT_cSC since they definitely show a Fermi surface.

The idea behind applying the Boson-Fermion scenario to HT_cSC is that we are confronted with inhomogeneous systems consisting of highly polarizable substructures on which localized bipolarons are formed. These local substructures are embedded in the rest of the lattice [5] which is occupied by electrons having a large either hole or electron-like Fermi surface [6,7], depending on doping. In such a two-component scenario the electrons scatter in a resonant fashion in and out of the Bosonic bipolaronic states. It is that which is at the origin of the opening of the pseudogap in the normal state of these materials, driven by a precursor of electron pairing [4], rather than magnetic interactions [6]. Generalizing this scenario in the way described above provides a mechanism by which the electrons acquire polaronic features (which, unlike for Bosons, are not of intrinsic nature) via the charge exchange term. This term thus not only controls the opening of the pseudogap as in the original BFM but also the appearance of the strong incoherent contributions to the electron spectrum arising from phonon shake-off effects. Given the two-subsystem picture on which the Boson-Fermion model is based, doping leads primarily to the creation of localized bipolarons which beyond a certain critical concentration are exchanged with the itinerant electrons. For a system such as, for instance, $YBCO$ the number $n_B = \langle b_i^+ b_i\rangle$ of doping induced bipolarons (approximately given by half the number of dopant $O_2^{2-}(1)$ ions in the chains) varies between 0 and 0.5 per effective site and the number of Fermions $n_F = \sum_\sigma \langle c_{i\sigma}^+ c_{i\sigma}\rangle$ is equal to 1 if the Boson-Fermion exchange coupling were absent. We thus obtain a total number of charge carriers $n_{tot} = n_F + 2n_B$ close to 2 for optimally doped systems. We should however remember that in real systems doping not only changes n_{tot} but also the relative occupancy of Fermions and Bosons, which seems to be the most important effect in the doping mechanism of these materials, achieved in our model as soon as v is different from zero.

We shall in the following solve the generalized BFM in the atomic limit (i.e., putting the second term in Eq.(2) equal to zero) for a grand canonical ensemble. In this case the eigenstates of the Hamiltonian are

$$\begin{aligned}
|0,l\rangle &= |\ 0\rangle \otimes |0) \otimes |\Phi(X)\rangle_l \\
|1,l\rangle &= |\uparrow\rangle \otimes |0) \otimes |\Phi(X)\rangle_l \\
|2,l\rangle &= |\downarrow\rangle \otimes |0) \otimes |\Phi(X)\rangle_l \\
|3,l\rangle &= u_{l,+}|\uparrow\downarrow\rangle \otimes |0) \otimes |\Phi(X)\rangle_{u_{l,+}} \\
&\qquad + v_{l,+}|0\rangle \otimes |1) \otimes |\Phi(X)\rangle_{v_{l,+}} \\
|4,l\rangle &= u_{l,-}|\uparrow\downarrow\rangle \otimes |0) \otimes |\Phi(X)\rangle_{u_{l,-}} \\
&\qquad + v_{l,-}|0\rangle \otimes |1) \otimes |\Phi(X)\rangle_{v_{l,-}} \\
|5,l\rangle &= |\uparrow\rangle \otimes |1) \otimes |\Phi(X - X_0)\rangle_l \\
|6,l\rangle &= |\downarrow\rangle \otimes |1) \otimes |\Phi(X - X_0)\rangle_l \\
|7,l\rangle &= |\uparrow\downarrow\rangle \otimes |1) \otimes |\Phi(X - X_0)\rangle_l \quad ,
\end{aligned} \tag{3}$$

where $|\sigma\rangle$ denotes a site occupied by an electron with spin σ and $|\uparrow\downarrow\rangle$ a site occupied by a pair of electrons with spin up and down. $|0)$ and $|1)$ denote a site unoccupied and, respectively, occupied by a Boson. $|\Phi(X)\rangle_l$ denotes the l-th excited oscillator state and $|\Phi(X - X_0)\rangle_l = (a^+ - \alpha)^l/\sqrt{l!}\, exp(\alpha(a - a^+))|\Phi(x)\rangle_0$ the l-th excited shifted oscillator state. These two sets of oscillator states are sufficient to describe all the states listed in Eq.(3) except for the states $|3,l\rangle$ and $|4,l\rangle$ for which the corresponding oscillator states are given by $|\Phi(X)\rangle_{u_{l,\pm}}$ and $|\Phi(X)\rangle_{v_{l,\pm}}$. The latter are determined by numerical diagonalization by expanding them in a set of excited harmonic oscillator states in the form $u_{l,\pm}|\Phi(X)\rangle_{u_{l,\pm}} = \sum_n u_{l,\pm}^n |\Phi(X)\rangle_n$ and $v_{l,\pm}|\Phi(X)\rangle_{v_{l,\pm}} = \sum_n v_{l,\pm}^n |\Phi(X)\rangle_n$. For the regime of coupling parameters which we are interested in we take into account up to 50 phonon states, i.e., $n \leq 50$. It is the states $|3,l\rangle$ and $|4,l\rangle$ which describe the transfer of polaronic features from the localized bipolarons to the conduction electrons when Boson-Fermion exchange processes take place. Since photoemission only couples to the electrons, it is via this transfer of polaronic features to the intrinsically non-polaronic electrons that photoemission spectra show features which are reminiscent of polaronic quasi-particles. These temporarily fluctuating local lattice deformations described by the corresponding oscillator wave functions $|\Phi(X)\rangle_{u_{l,\pm}}$ and $|\Phi(X)\rangle_{v_{l,\pm}}$ are manifest in the pair distribution function (PDF)

$$g(x) = \frac{1}{Z}\sum_{n,l} \exp(-\beta E(n,l))\langle n,l|\delta(x)|n,l\rangle \quad . \tag{4}$$

Here $Z = \sum_{m=0}^{7} \sum_{l=0}^{\infty} e^{-\beta E(m,l)}$ denotes the partition function, with $E(m,l)$ being the eigenvalues of the eigenstates listed above, given by: $E(0,l) = l\hbar\omega_0$, $E(1,l) = E(2,l) = \varepsilon_0 + l\hbar\omega_0$, $E(3,l) = \varepsilon_{l,+}$, $E(4,l) = \varepsilon_{l,-}$, $E(5,l) = E(6,l) = \varepsilon_0 + E_0 + l\hbar\omega_0 - \varepsilon_{BP}$ and $E(7,l) = 2\varepsilon_0 + E_0 + l\hbar\omega_0 - \varepsilon_{BP}$, with $\varepsilon_0 = D - \mu$ and $E_0 = \Delta_B - 2\mu$.

In order to investigate the various physical quantities on the basis of this single-site generalized BFM we must choose Δ_B in a way to guarantee the conditions set out above, that is, a concentration of electrons $n_F \simeq 0.75$ (corresponding to a hole concentration of $\simeq 0.25$) for a total concentration of particles $n_{tot} = 2$. In order to achieve these conditions we put the bare bipolaronic level Δ_B above the bare electronic energy level D such that the bipolaronic levelshift ε_{BP} brings this level down slightly below the bare electronic level. We adjust the precise position of this level by putting $\Delta_B = 2D + \hbar\omega_0\alpha^2 - \delta\Delta_B$, with $\delta\Delta_B = 0.025$, chosen in order to give $n_F \simeq 0.75$.

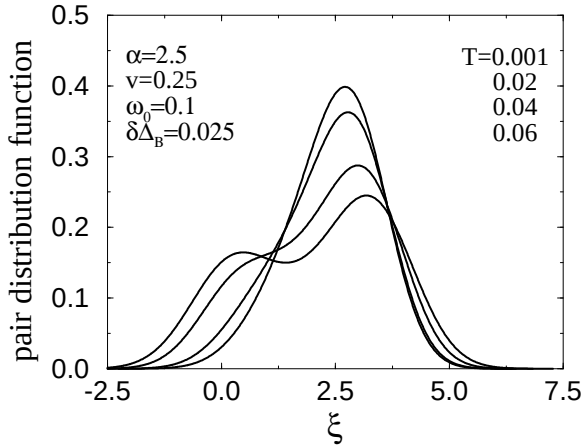

FIG. 1. The pair distribution function for several temperatures T in units of D, the highest T corresponding to the most pronounced double-peak structure. The displacement is measured by the dimensionless parameter $\xi = X\sqrt{M\omega_0/\hbar}$.

Given this choice of parameters, we obtain a PDF (illustrated in Fig.1) showing a double-peak structure which merges into a single-peak structure as the temperature is lowered below the characteristic temperature T^*, at which, as we shall see below, the pseudogap opens up. The two peak positions characterize the two deformations of the local lattice environment where a given site is alternatively occupied by a pair of electrons or by a bipolaron. Recent $EXAFS$ [5], $XANES$ [8] and pulsed neutron scattering [9] experiments give some indications for such dynamical local lattice fluctuations.

Let us now embark on the evaluation of the intensity of the photoemission spectrum $I_{PES}(\omega)$ from a single site Boson-Fermion system - tantamount to the angle integrated rather than angle resolved photoemission spectroscopy when neglecting the effect of the dynamical mean field coming from the itinerancy of the electrons. We have $I_{PES}(\omega) = I_E(\omega) n_F(\omega)$, where $n_F(\omega)$ denotes the Fermi distribution function and $I_E(\omega)$ the emission part of the total one-particle Fermionic spectral function

$$
\begin{aligned}
I(\omega) &= \frac{1}{Z} \sum_{m,m';l,l'} \left(e^{-\beta E(m,l)} + e^{-\beta E(m',l')} \right) \\
&\quad |\langle m', l'|c_\uparrow|m, l\rangle|^2 \delta(\omega - E(m,l) + E(m',l')) \\
&= Z_F \delta(\omega - \varepsilon_0) + \frac{1}{Z} \sum_{l,m,s=\pm} |u_{l,s}^m|^2 (e^{-\beta(\varepsilon_0 + m\hbar\omega_0)} \\
&\quad + e^{-\beta\varepsilon_{l,s}}) \delta(\omega + \varepsilon_0 + m\hbar\omega_0 - \varepsilon_{l,s}) \\
&+ \frac{e^{-\alpha^2}}{Z} \sum_{l,m,s=\pm} \left| \sum_{n \le l} v_{m,s}^n \sqrt{\frac{l!}{n!}} \sum_{n'=0}^{n} \binom{n}{n'} \frac{\alpha^{n-n'}(-\alpha)^{l-n'}}{(l-n'!)} \right|^2 \\
&(e^{-\beta\varepsilon_{m,s}} + e^{-\beta(\varepsilon_0 + E_0 + l\hbar\omega_0 - \varepsilon_{BP})}) \delta(\omega + \varepsilon_{m,s} - \varepsilon_0 - E_0 \\
&- l\hbar\omega_0 + \varepsilon_{BP}) \quad . \qquad (5)
\end{aligned}
$$

Here $Z_F = \frac{1}{Z}(1 + e^{-\beta\varepsilon_0})(1 + e^{-\beta(\varepsilon_0 + E_0 - \varepsilon_{BP})}) n_B(\hbar\omega_0)$ represents the spectral weight of the non-bonding contributions which accounts for the coherent part of the photoemission spectrum, unaffected by any coupling to the Bosons and hence to the phonons ($n_B(\omega)$ denotes the Bose distribution function). The second and third contribution to the spectral function $I(\omega)$ account for the incoherent part of the spectrum. We illustrate in Fig.2 the photoemission spectral intensity $I_{PES}(\omega)$ for different temperatures (in units of D). For high temperatures ($T \simeq 0.06$) we observe a very much broadened spectral function which in shape comes close to that of a typical Fermi liquid. Upon lowering the temperature this spectral function starts exhibiting a pseudogap and at the same time a broad incoherent contribution (coming from the second and third term of the expression for $I(\omega)$ in Eq.(5)) emerges. The incoherent part of the spectrum extends over a region in energy which is of the order of the half band width ($\simeq 0.5eV$) and is practically temperature independent at low temperatures, which seems to be confirmed experimentally [10]. The closing up of the pseudogap (measured as the difference in energy between the chemical potential at $\omega = 0$ and the midpoint of the leading edge of the photoemission spectrum) as we increase the temperature is illustrated in the inset of Fig.2. The pseudogap has a zero temperature limit of $0.085D \simeq 40\,meV$ and closes up at a characteristic temperature $T^* \simeq 0.06D \simeq 350\,K$, which are reasonable numbers. We should add that the chemical potential for temperatures below T^* turns out to be practically temperature independent. In order to illustrate the closing up of the pseudogap as the temperature approaches T^* we plot in Fig.3 the density of states $I(\omega)$ for different temperatures. We clearly notice a strongly non-symmetric bias near the chemical potential ($\omega = 0$) which seems to be verified in tunneling experiments [11].

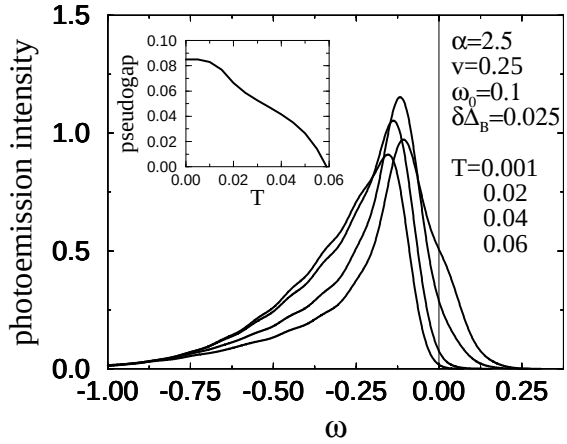

FIG. 2. The photoemission spectral intensity $I_{PES}(\omega)$, taking into account experimental broadening of $\Delta\omega = 0.05$, for various temperatures T, the lowest T corresponding to the curves with the largest pseudogap. The inset shows the temperature behaviour of the pseudogap.

The work reported in this Letter relates the temperature dependence of the pseudogap to that of the incoherent part of the quasi-particle spectrum. The opening of the pseudogap, being associated to resonant exchange tunneling between intrinsically unpaired electrons and electrons paired up in bipolaronic states, is thus driven by a metal-insulator cross-over rather than by superconducting fluctuations and thus can open up well above the onset of the superconducting phase. The broad incoherent part of this spectrum is attributed to phonon shake-off effects arising from the polaronic character of the electrons in the metallic layers, transmitted to them via their resonant scattering into bipolaronic states. The temporarily fluctuating local lattice deformations which are caused in this process are expected to show up in a characteristic double-peak feature of the pair distribution function (measureable by $EXAFS$) and should test whether the incoherent $ARPES$ background is of polaronic origin or not. Our approach is based on an atomic limit calculation of the generalized Boson-Fermion model which is solved exactly by numerical means. The results obtained are not expected to change qualitatively when taking into account the itinerancy of the electrons. This will only introduce possible asymmetries in the Brillouin zone coming from asymmetric coupling v in a more microscopic model (in accordance with the hypothesis of strongly hybridized plane and out of plane states in certain parts of the Brilloun zone, as suggested by LDA calculations [12]) and affect the quasi-particle structure close to the Fermi energy. For this energy regime our self-consistent studies [4] on the original BFM, fully taking into account electron itinerancy but neglecting any coupling to the phonons, reproduces more faithfully the quasi particle structure, but with an incoherent contribution which is typically of the order of 0.1 D. This is one order of magnitude less than the incoherent contributions due to phonon shake-off reported in this Letter. We therefore can safely treat this problem within the approximation scheme presented here. Our results are moreover robust in the sense that they hold for different total concentrations between 1.5 and 2, as long as we enforce the condition that n_F is between 0.7 and 1.

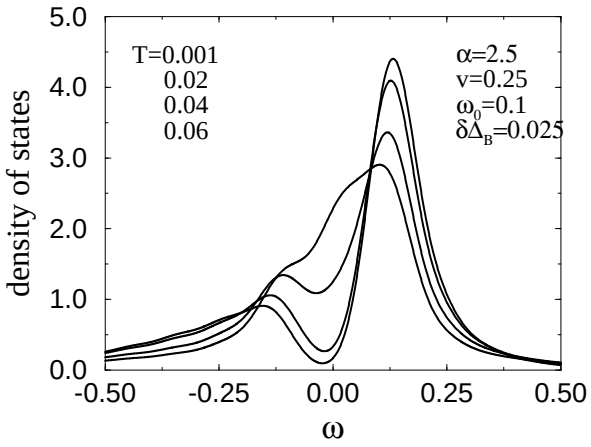

FIG. 3. Evolution of the electron DOS $I(\omega)$ as a function of temperature T, the lowest T corresponding to the deepest pseudogap (experimental broadening $\Delta\omega = 0.05$).

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[12] see for instance O.K. Andersen et al., Phys. Rev. B **49**, 4145 (1994) for $YBCO$. Similar results were obtained for Hg, Tl and Bi compounds.